

Age of Meteorites by the  $A^{40}-K^{40}$  Method\*G. J. WASSERBURG, *Institute for Nuclear Studies, University of Chicago, Chicago, Illinois*

AND

R. J. HAYDEN, *Argonne National Laboratory, Lemont, Illinois*

(Received August 11, 1954; revised manuscript received September 29, 1954)

The ages of two stony meteorites were determined by measuring the  $A^{40}/K^{40}$  ratio using an isotopic dilution technique. With a branching ratio  $\lambda_e/\lambda_\beta=0.085$  and a decay constant  $\lambda=0.55\times 10^{-9}$  yr $^{-1}$ , the ages obtained were  $(4.82\pm 0.20)\times 10^9$  and  $(4.58\pm 0.20)\times 10^9$  years.

THE radiogenic argon content of two stony meteorites was determined using an isotopic dilution technique. These meteorites were Beardsley, a grey chondrite, and Forest City, a brecciated spherical bronzite chondrite.<sup>1</sup> The extraction procedure used has been described previously.<sup>2</sup> The only modification of the procedure was that a CuO furnace at 500°C was attached directly to the reaction vessel. This was necessary in order to convert the large quantity of hydrogen, which was produced in the reaction of the metallic iron in the meteorite with the NaOH flux, into water. Additional experiments with mixtures of feldspar and metallic iron indicate that the continuous reaction of the hydrogen does not effect the mixing of tracer and evolved argon. Two sets of  $A^{38}$  tracers were used, each of which contained better than 80 percent  $A^{38}$ . The amount of radiogenic argon was computed from data given in Tables I and II. The correction for normal argon contamination was made assuming that the mass 36 peak was due to atmospheric argon. The discrimination of the mass spectrometer was determined by running normal argon of spectroscopic purity before and after each sample. Nier's values for the isotopic composition of normal argon were assumed.<sup>3</sup> No hydrocarbons were observed in the mass 36 to 40 region. In the first Beardsley experiment the HCl correction to the  $A^{36}$  peak resulted in a 1 percent change in the amount of radiogenic  $A^{40}$ . In the second Beardsley and Forest City experiments, the HCl correction resulted in 0.4 percent and 0.0 percent corrections, respectively, to the amount of radiogenic  $A^{40}$ .

In the second Beardsley experiment, a considerable portion of the material splattered to the cooler part of the reaction vessel and was unreacted. The amount of hydrogen and water produced during the reaction was much less than in the other two experiments.

The potassium determination was made using an isotopic dilution technique.  $K^{41}$  obtained from Oak Ridge was used as a tracer. The  $K^{39}/K^{41}$  ratio in the tracer was 0.00922.

\* This work was supported in part by a U. S. Atomic Energy Commission contract and in part by a National Science Foundation grant.

<sup>1</sup> Meteorites were obtained from H. H. Nininger, Sedona, Arizona.

<sup>2</sup> G. J. Wasserburg and R. J. Hayden, *Phys. Rev.* **93**, 645 (1954).

<sup>3</sup> A. O. Nier, *Phys. Rev.* **77**, 789 (1950).

Two aliquots of Beardsley meteorite and two aliquots of Forest City meteorite were analyzed for potassium. Two synthetic standards, one composed of a mixture of feldspar containing 10.43 percent potassium and olivine (containing less than 0.001 percent potassium) and the other a feldspar containing 8.06 percent potassium were analyzed along with the meteorites. The samples were dissolved in HF and H<sub>2</sub>SO<sub>4</sub>, and aliquots of the  $K^{41}$  tracer solution added. SiF<sub>4</sub> was removed by evaporation, and HCl was added. A potassium-rich filtrate was obtained by precipitating Al, Fe, Mg, and Ca with NH<sub>4</sub>OH and (NH<sub>4</sub>)<sub>2</sub>CO<sub>3</sub> solutions.

The isotopic composition was determined with a surface ionization technique using an oxidized tungsten filament. The samples were run on a mass spectrometer with a 12-inch radius of curvature and 60° deflection. The discrimination of the instrument was determined by running a mixture of tracer  $K^{41}$  and normal K and also by running a sample of normal potassium. By assuming Nier's value for the isotopic ratio in normal potassium<sup>3</sup> a value for the discrimination was obtained which agrees within 1.5 percent with the value obtained from the tracer—normal mixture. It was assumed that the isotopic composition of the meteoritic potassium was normal.

By assuming a decay constant  $\lambda=0.55\times 10^{-9}$ /yr and a branching ratio  $\lambda_e/\lambda_\beta=0.085$ ,<sup>4</sup> the following ages are calculated: Beardsley I,  $(4.82\pm 0.20)\times 10^9$  yr; Beardsley II,  $(4.50\pm 0.20)\times 10^9$  yr; Forest City,  $(4.67\pm 0.20)\times 10^9$  yr. The lower age obtained for Beardsley II is believed to be due to the incomplete reaction discussed earlier.

The branching ratio which was used in the calculation was determined by the authors by determining the  $A^{40}/K^{40}$  ratio and the  $Pb^{206}/U^{238}$ ,  $Pb^{207}/U^{235}$  ages of several coexisting potassium and uranium minerals from over a considerable span of geologic time.<sup>4</sup> The value for  $\lambda$  given above was assumed in the branching ratio calculations. The values given for the error in the ages were obtained by assuming a 3 percent error in  $\lambda$  and 6 percent error in  $\lambda/\lambda_e(A^{40}/K^{40})$ .

Previous measurements of the age of meteorites by

<sup>4</sup> G. J. Wasserburg and R. J. Hayden, Presented at the May, 1954, meeting of the American Geophysical Union.

Gerling *et al.*<sup>5</sup> using the A<sup>40</sup>-K<sup>40</sup> method gave values ranging from  $0.5 \times 10^9$  yr to  $3.03 \times 10^9$  yr. These values were computed by using a decay constant  $\lambda = 0.55 \times 10^{-9}$  yr<sup>-1</sup> and a branching ratio  $\lambda_e/\lambda_\beta = 0.124$ . Recomputing their data using a branching ratio of 0.085, we obtain an age for their oldest sample of  $3.55 \times 10^9$  yr. The A<sup>40</sup>/K<sup>40</sup> ratios which we obtain differ from Gerling's largest value by a factor of 2.2.

Paneth *et al.*<sup>6</sup> have found that a considerable portion of the helium contained in iron meteorites was produced by cosmic radiation. The amount of helium found by Paneth *et al.* in iron meteorites is less than  $4 \times 10^{-5}$  cc STP per gram. If it is assumed that the iron meteorites have retained most of the helium produced in them by cosmic radiation, and that they are of the same age as the stony meteorites, one can calculate the effect of the cosmic-ray products on the A<sup>40</sup> age. As the stony meteorites contain approximately 25 percent iron, the amount of helium produced from this iron is about  $10^{-5}$  cc STP per gram. If the same amount of A<sup>40</sup> were produced this would have a 2 percent effect on the A<sup>40</sup>-K<sup>40</sup> age. This estimate of the amount of argon produced by cosmic radiation on iron is certainly much too high.

TABLE I. Argon analysis.

Sample	Sample wt (g)	A <sup>38</sup> tracer (cc STP)	A <sup>36</sup> /A <sup>38</sup>	A <sup>38</sup> /A <sup>40</sup>	Radio-genic A <sup>40</sup> (cc STP) per g
Beardsley I	79.485	$3.74 \times 10^{-5}$	0.0383	$0.00623 \pm 0.00004$	$7.13 \times 10^{-5}$
Beardsley II	29.862	$8.31 \times 10^{-5}$	0.0236	$0.0358 \pm 0.0002$	$5.87 \times 10^{-5}$
Forest City	71.762	$8.72 \times 10^{-5}$	0.0227	$0.01987 \pm 0.00004$	$5.34 \times 10^{-5}$

<sup>5</sup> E. K. Gerling and T. G. Pavlova, Doklady Akad. Nauk S.S.S.R. **77**, No. 1, 85 (1951); E. L. Krinov, Priroda Izd. Akad. Nauk S.S.S.R., Moskva **40**, No. 10, 83, (1951).

<sup>6</sup> F. A. Paneth and K. I. Mayne, Nature **172**, 200 (1953), F. A. Paneth *et al.*, Geochim. et Cosmochim. Acta **3**, 257 (1953).

TABLE II. Potassium analysis.

Sample	Sample wt (g)	K tracer added (mg)	K <sup>41</sup> /K <sup>39</sup>	% K (by wt)
Beardsley A	3.600	0.792	$0.294 \pm 0.003$	0.101
Beardsley B	1.980	0.792	$0.474 \pm 0.007$	0.101
Standard (0.104% K)	10.455	1.584	$0.214 \pm 0.004$	0.110
Forest City A	2.317	0.792	$0.492 \pm 0.006$	0.0827
Forest City B	1.015	0.792	$1.016 \pm 0.010$	0.0835
Standard (8.06% K)	0.1243	1.584	$0.231 \pm 0.002$	8.25

If A<sup>36</sup>, A<sup>38</sup>, and A<sup>40</sup> are produced equally to within two orders of magnitude, then the effect of cosmic-ray produced argon is to make the computed age less than the true age. If it is also true that the He/A ratio in spallation products from iron is 1000 or greater the effect on the age is negligible.

The authors have found no evidence for the loss of argon by diffusion in terrestrial feldspars,<sup>7</sup> but it is possible that the meteorites have lost some argon by this means. Hence the values reported are interpreted as lower limits to the age of the samples. If it is assumed that the meteorites are cogenetic with the solar system, the above value may also be applied as a lower limit to the age of the solar system.

The value of  $4.8 \times 10^9$  years obtained for Beardsley is a lower limit to the age of the universe. This value is in accord with the lower limit of  $4.5 \times 10^9$  yr calculated by Patterson<sup>8</sup> from the isotopic composition of modern terrestrial lead and meteoritic lead.

## ACKNOWLEDGMENT

The authors are greatly indebted to Professor M. G. Inghram and to Professor H. C. Urey for their valuable suggestions during the progress of this work.

<sup>7</sup> G. J. Wasserburg and R. J. Hayden, Geochim. et Cosmochim. Acta (to be published).

<sup>8</sup> C. Patterson, *Nuclear Processes in Geologic Settings* (National Research Council, Washington, D. C., 1953), p. 36.